Giant Anharmonicity and Electron-Phonon Mediated Superconductivity in MgB_2 at 39 K

The recent surprise discovery of superconductivity in MgB_2 at 39 K has stimulated a great deal of research on this intercalated graphite-like system (see Fig. 1). Sparked by this discovery, we set out to unlock the structural secrets and, in particular, to reveal the origin of the high T_c in MgB_2: an electron-phonon or other exotic mechanism? To answer this fundamental question, we calculated T_c, its pressure dependence, and its isotope effect from the electronic band structure and lattice dynamics of MgB_2 using density functional theory within the generalized gradient approximation [1–3].

Figure 2 shows that the features in the calculated phonon density of states (DOS) are in excellent agreement with the neutron data (GDOS), giving confidence that the calculations provide a sound description of the physical properties of the system. The DOS consists of two bands of phonons, one below 40 meV corresponding primarily to acoustic phonon modes, and one above 50 meV that mostly involves the boron motions. Inspection of the calculated phonon-dispersion curves that make up the high-energy band in the DOS reveals that the in-plane boron phonons (as depicted in the inset to Fig. 3) are very anharmonic. To demonstrate this, in Fig. 3 we plot the total energy as boron atoms move according to one of these zone-center in-plane phonons with E_{2g} symmetry. The potential indicates a very strong anharmonic term. Numerically solving the Schrödinger equation for this anharmonic potential yields a phonon energy of $\hbar\omega(E_{2g})$ = 74.5 meV, a 25 % enhancement over the harmonic value of 60.3 meV. This value is in good agreement with recent Raman measurements. The giant anharmonicity revealed gives the first hint that the in-plane modes are strongly coupled to the p_{xy} σ bonding orbitals of boron, as shown schematically in Fig.1. This coupling is also evident from the splitting of the boron σ bands (red lines) with the E_{2g} phonons (see Fig. 3). Note that the other bands are not affected by the E_{2g} phonons.

The splitting of the boron σ bands, when averaged over the Fermi surface, gives an electron-phonon (EP) coupling constant $\lambda \approx 1$. Using this value in the McMillan expression for T_c with $\omega(E_{2g})$ and taking a typical value for the Coulomb repulsion $\mu^* = 0.15$, we obtain a T_c of 39.4 K, in excellent agreement with experiments. We also solved the Schrödinger equation for the potential shown in Fig. 3 for different boron masses and obtained $\omega(E_{2g}) = 291.8\, M^{-0.575}$ and $\lambda = 0.6151\, M^{0.169}$, which yields the T_c-M curve shown in Fig. 4 and a boron isotope effect $\alpha = 0.21$, in good agreement with the experimental value of 0.26 ± 0.03.

Since the pressure dependence of T_c puts a stringent test on any theory of superconductivity, we repeated the calculations of phonons and electronic band structures discussed above for isotropic, uniaxial (along *c*-axis), and biaxial (in the *ab*-plane) pressures [3]. We find that while

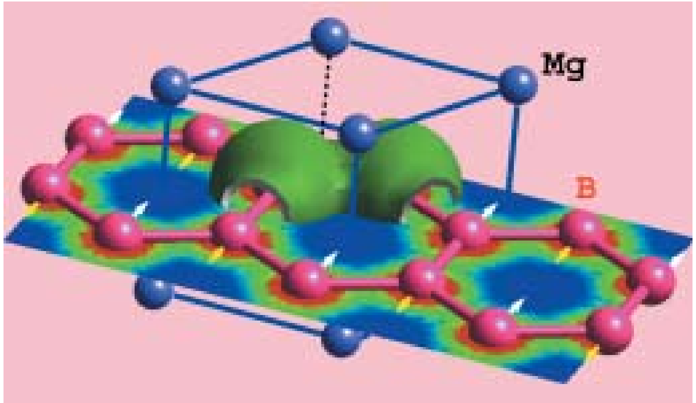

FIGURE 1. The crystal structure of MgB_2 consisting of B and Mg hexagonal layers. The in-plane boron modes (shown by arrows) are strongly coupled to the boron $p_{x,y}$ σ bands shown as the green contour and isosurface plots.

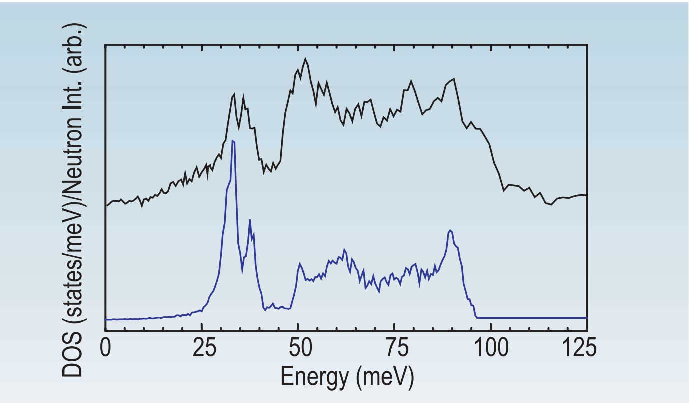

FIGURE 2. Generalized (top) and the calculated (bottom) phonon density of states. The intensities of the peaks do not agree well because in DOS B and Mg ions contribute equally while in GDOS they are weighted by neutron cross-sections and inverse masses.

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$\omega(E_{2g})$ increases with increasing pressure, the density of states at the Fermi energy decreases. The EP constant λ also shows significant changes with pressure. Inserting all these competitive effects into the McMillan formula yields the pressure dependence of T_c shown in Fig. 4.

For isotropic pressure T_c decreases with increasing pressure almost linearly at a rate of ≈ -1.0 K/GPa, in excellent agreement with the experimental value of -1.1 K/GPa. We also predict a cusp in the T_c-P curve around $P \approx 20$ GPa.

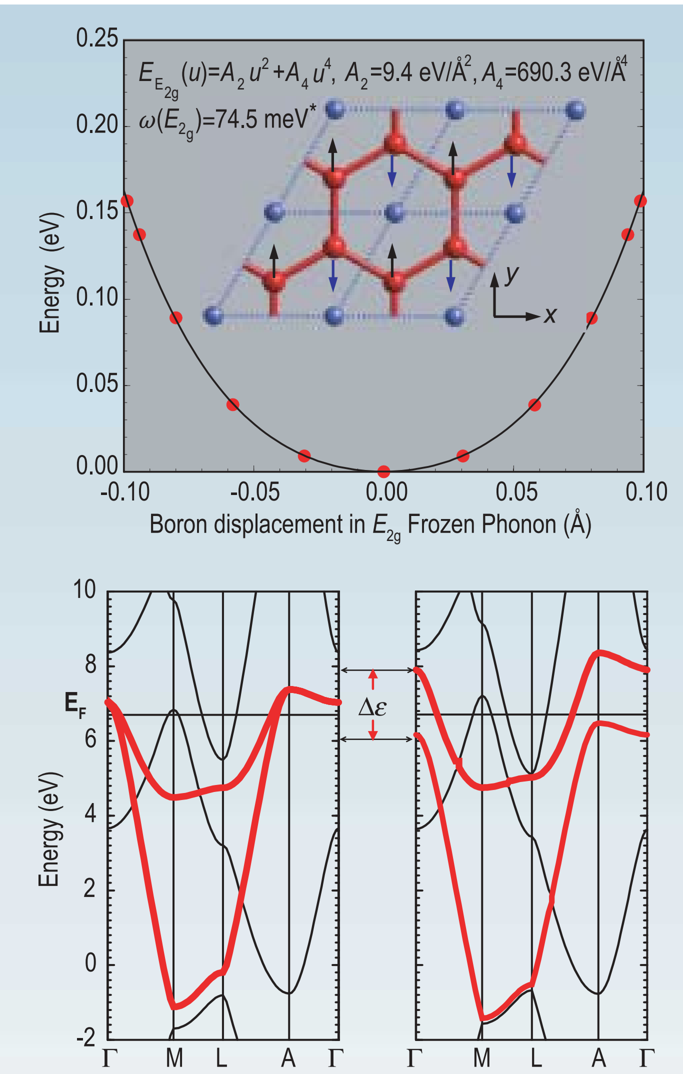

FIGURE 3. Top: Total energy curve as a function of boron displacement for E_{2g} mode (shown in the inset), indicating a large anharmonic term in the potential. Bottom: Band structure of the undistorted (left) and distorted structures (right) by E_{2g} phonons ($u_B \approx 0.06$ Å). See Ref. 1 for the animation of the zone center phonons and their coupling with the B σ bands.

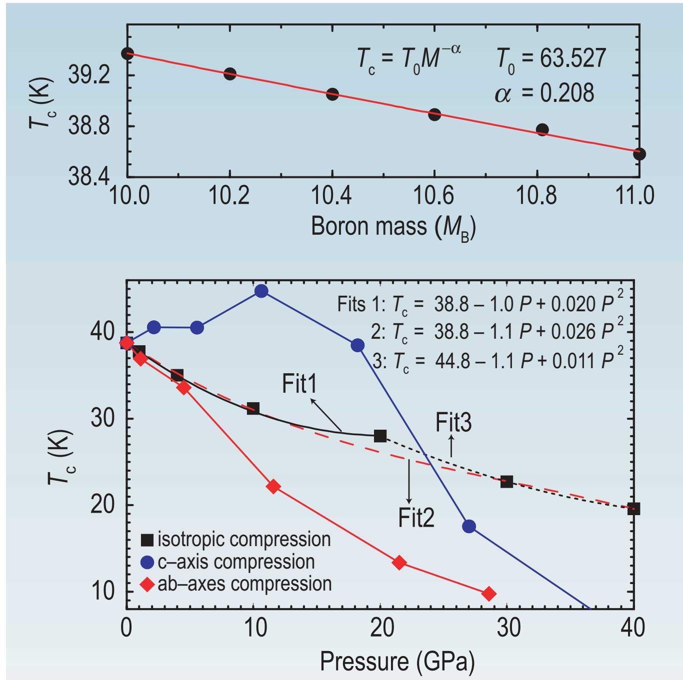

FIGURE 4. Top: Boron mass dependence of the T_c. Bottom: Pressure dependence of T_c as a function of uniform *ab*-axes and *c*-axis compression, respectively.

A similar cusp was recently observed experimentally. Our calculations indicate that T_c should increase first and then decrease with increasing *c*-compression, while it should decrease rapidly with *ab*-compression. Hence, when single crystal samples of MgB_2 become available, measurements of the *ab*- and *c*-compression dependence of T_c should provide a critical test of our theory.

MgB_2 may be the ultimate BCS s-wave superconductor, with parameters controlling T_c fully optimized to yield the highest possible T_c. However, even if T_c cannot be increased further, the low cost, light mass, easy fabrication, nearly isotropic high conductivity of MgB_2, which also has a large critical current, will no doubt find many important technological applications in the near future.

References

[1] For full details of this work, see the website: http://www.ncnr.nist.gov/staff/taner/mgb2.

[2] T. Yildirim, O. Gulseren, J. F. Lynn, C. M. Brown, T. J. Udovic, Q. Huang, N. Rogado, A. Regan, M. A. Hayward, J. S. Slusky, T. He, M. K. Haas, P. Khalifah, K. Inumaru, and R. J. Cava, Phys. Rev. Lett. **87**, 37001 (2001).

[3] T. Yildirim and O. Gulseren, J. Phys.Chem. Solids (in press, 2002).